

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

APPLICANT: R. Formato, et al.

SERIAL NO. Not Yet Assigned EXAMINER: Not Yet Assigned
[Divisional Application of
U.S.S.N. 09/261,349, filed 3/3/99
Express Mail Label No. EL298355479US]

FILED: Herewith GROUP: Not Yet Assigned

FOR: COMPOSITE SOLID POLYMER ELECTROLYTE MEMBRANES

ASSISTANT COMMISSIONER FOR PATENTS
WASHINGTON, DC 20231

Sir:

PRELIMINARY AMENDMENT

Kindly amend the above-identified application as follows:

IN THE SPECIFICATION

Page 29, Table 3, delete the second line of data in its entirety, which line reads:

" Resistivity	14.7	11.38	9.5 - 10.0".
(ohm-cm)			

Page 36, Table 8, second line of data corresponding to sample FMI 539-22-1, delete the Resistance data appearing in column 8, "0.056", and insert therefor --NT--.

Page 36, Table 8, third line of data corresponding to sample FMI 539-22-2, delete the Resistance data appearing in column 8, "0.043", and insert therefor --NT--.

IN THE CLAIMS

Please cancel claims 1-50.

Please add the following new claims.

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51. (new) A method of producing a composite solid polymer electrolyte membrane (SPEM) comprising a porous polymer substrate interpenetrated with an ion-conducting material, said method comprising the steps of preparing a mixture of the polymer substrate and the ion-conducting material and casting or extruding the composite SPEM from the mixture, and wherein

(i) the porous polymer substrate comprises a homopolymer or copolymer of a liquid crystalline polymer or a solvent soluble thermoset or thermoplastic aromatic polymer; and

(ii) the ion-conducting material comprises a homopolymer or copolymer of at least one of a sulfonated, phosphonated or carboxylated ion-conducting aromatic polymer or a perfluorinated ionomer.

52. (new) The method of claim 51, wherein the SPEM is substantially thermally stable to temperatures of at least about 100°C.

53. (new) The method of claim 51, wherein the SPEM is substantially thermally stable from at least about 100°C to at least about 175°C.

54. (new) The method of claim 51, wherein the liquid crystalline polymer substrate comprises a lyotropic liquid crystalline polymer.

55. (new) The method of claim 54, wherein the lyotropic liquid crystalline polymer substrate comprises at least one of a polybenzazole (PBZ) and polyaramid (PAR) polymer.

56. (new) The method of claim 55, wherein the polybenzazole polymer substrate comprises a homopolymer or copolymer of at least one of a polybenzoxazole (PBO), polybenzothiazole (PBT) and polybenzimidazole (PBI) polymer and the

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polyaramid polymer comprises a homopolymer or copolymer of a polypara-phenylene terephthalamide (PPTA) polymer.

57. (new) The method of claim 51, wherein the thermoset or thermoplastic aromatic polymer substrate comprises at least one of a polysulfone (PSU), polyimide (PI), polyphenylene oxide (PPO), polyphenylene sulfoxide (PPSO), polyphenylene sulfide (PPS), polyphenylene sulfide sulfone (PPS/SO₂), polyparaphenylene (PPP), polyphenylquinoxaline (PPQ), polyaryleketone (PK) and polyetherketone (PEK) polymer.

58. (new) The method of claim 57, wherein the polysulfone polymer substrate comprises at least one of a polyethersulfone (PES), polyetherethersulfone (PEES), polyarylethersulfone (PAS), polyphenylsulfone (PPSU) and polyphenylene sulfone (PPSO₂) polymer; the polyimide (PI) polymer comprises a polyetherimide (PEI) polymer; the polyetherketone (PEK) polymer comprises at least one of a polyetherketone (PEK), polyetheretherketone (PEEK), polyetherketone-ketone (PEKK), polyetheretherketone-ketone (PEEKK) and polyetherketoneetherketone-ketone (PEKEKK) polymer; and the polyphenylene oxide (PPO) polymer comprises a 2,6-diphenyl PPO or 2,6 dimethyl PPO polymer.

59. (new) The method of claim 51, wherein the ion-conducting aromatic polymer comprises a wholly aromatic ion-conducting polymer.

60. (new) The method of claim 51, wherein the ion-conducting aromatic polymer comprises a sulfonated, phosphonated or carboxylated polyimide polymer.

61. (new) The method of claim 60, wherein the polyimide polymer is fluorinated.

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62. (new) The method of claim 59, wherein the wholly-aromatic ion-conducting polymer comprises a sulfonated derivative of at least one of a polysulfone (PSU), polyphenylene oxide (PPO), polyphenylene sulfoxide (PPSO), polyphenylene sulfide (PPS), polyphenylene sulfide sulfone (PPS/SO₂), polyparaphenylene (PPP), polyphenylquinoxaline (PPQ), polyarylsulfone (PK), polyetherketone (PEK), polybenzazole (PBZ) and polyaramid (PAR) polymer.

63. (new) The method of claim 62, wherein:

(i) the polysulfone polymer comprises at least one of a polyethersulfone (PES), polyetherethersulfone (PEES), polyarylsulfone, polyarylethersulfone (PAS), polyphenylsulfone (PPSU) and polyphenylenesulfone (PPSO₂) polymer,

(ii) the polybenzazole (PBZ) polymer comprises a polybenzoxazole (PBO) polymer;

(iii) the polyetherketone (PEK) polymer comprises at least one of a polyetherketone (PEK), polyetheretherketone (PEEK), polyetherketone-ketone (PEKK), polyetheretherketone-ketone (PEEKK) and polyetherketoneetherketone-ketone (PEKEKK) polymer; and

(iv) the polyphenylene oxide (PPO) polymer comprises at least one of a 2,6-diphenyl PPO, 2,6-dimethyl PPO and 1,4-poly phenylene oxide polymer.

64. (new) The method of claim 51, wherein the perfluorinated ionomer comprises a homopolymer or copolymer of a perfluorovinyl ether sulfonic acid.

65. (new) The method of claim 64, wherein the perfluorovinyl ether sulfonic acid is carboxylic- (COOH), phosphonic- (PO(OH)₂) or sulfonic- (SO₃H) substituted.

66. (new) The method of claim 51, wherein the ion-conducting material comprises at least one of a polystyrene sulfonic acid (PSSA), poly(trifluorostyrene)

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sulfonic acid, polyvinyl phosphonic acid (PVPA), polyacrylic acid and polyvinyl sulfonic acid (PVSA) polymer.

67. (new) The method of claim 51, wherein the porous polymer substrate comprises a homopolymer or copolymer of at least one of a substituted or unsubstituted polybenzazole polymer, and wherein the ion-conducting material comprises a sulfonated derivative of a homopolymer or copolymer of at least one of a polysulfone (PSU), polyphenylene sulfoxide (PPSO) and polyphenylene sulfide sulfone (PPS/SO₂) polymer.

68. (new) The method of claim 67, wherein the polysulfone polymer comprises at least one of a polyethersulfone (PES) and polyphenylsulfone (PPSU) polymer.

69. (new) The method of claim 51, further comprising cross-linking the ion-conducting material to form sulfone crosslinkages.

70. (new) The method of claim 51, further comprising chlorinating or brominating the ion-conducting material.

71. (new) The method of claim 51, further comprising adding antioxidants to the ion-conducting material.

72. (new) The method of claim 51, further comprising purifying the ion-conducting material.

73. (new) The method of claim 72, wherein purifying the ion-conducting material comprises dissolving the ion-conducting material in a suitable solvent and precipitating the ion-conducting material into a suitable non-solvent.

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74. (new) The method of claim 72, wherein purifying the ion-conducting material comprises substantially removing overly sulfonated or degraded fractions of the ion-conducting material.

75. (new) The method of claim 51, wherein the mixture of the polymer substrate and ion-conducting material is prepared in a common solvent.

76. The method of claim 75, wherein the common solvent is selected from the group consisting of tetrahydrofuran (THF), dimethylacetamide (DMAc), dimethylformamide (DMF), dimethylsulfoxide (DMSO), N-Methyl-2-pyrrolidinone (NMP), sulfuric acid, phosphoric acid, chlorosulfonic acid, polyphosphoric acid (PPA) and methanesulfonic acid (MSA).

77. (new) A method of producing a composite solid polymer electrolyte membrane (SPEM), said method comprising the steps of performing a sulfonation reaction within the pores of a polymer substrate, wherein the polymer substrate comprises a homopolymer or copolymer of a liquid crystalline polymer or a solvent soluble thermoset or thermoplastic aromatic polymer.

78. (new) The method of claim 77, wherein the SPEM is substantially thermally stable to temperatures of at least about 100°C.

79. (new) The method of claim 77, wherein the SPEM is substantially thermally stable from at least about 100°C to at least about 175°C.

80. (new) The method of claim 77, wherein the liquid crystalline polymer substrate comprises a lyotropic liquid crystalline polymer.

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81. (new) The method of claim 80, wherein the lyotropic liquid crystalline polymer substrate comprises at least one of a polybenzazole (PBZ) and polyaramid (PAR) polymer.

82. (new) The method of claim 81, wherein the polybenzazole polymer substrate comprises a homopolymer or copolymer of at least one of a polybenzoxazole (PBO), polybenzothiazole (PBT) and polybenzimidazole (PBI) polymer and the polyaramid polymer comprises a homopolymer or copolymer of a polypara-phenylene terephthalamide (PPTA) polymer.

83. (new) The method of claim 77, wherein the thermoset or thermoplastic aromatic polymer substrate comprises at least one of a polysulfone (PSU), polyimide (PI), polyphenylene oxide (PPO), polyphenylene sulfoxide (PPSO), polyphenylene sulfide (PPS), polyphenylene sulfide sulfone (PPS/SO₂), polyparaphenylene (PPP), polyphenylquinoxaline (PPQ), polyaryletherone (PEK) and polyetheretherone (PEEK) polymer.

84. (new) The method of claim 83, wherein the polysulfone polymer substrate comprises at least one of a polyethersulfone (PES), polyetherethersulfone (PEES), polyarylethersulfone (PAS), polyphenylsulfone (PPSU) and polyphenylenesulfone (PPSO₂) polymer; the polyimide (PI) polymer comprises a polyetherimide (PEI) polymer; the polyetherketone (PEK) polymer comprises at least one of a polyetherketone (PEK), polyetheretherketone (PEEK), polyetherketone-ketone (PEKK), polyetheretherketone-ketone (PEEKK) and polyetherketoneetherketone-ketone (PEKEKK) polymer; and the polyphenylene oxide (PPO) polymer comprises a 2,6-diphenyl PPO or 2,6 dimethyl PPO polymer.

85. (new) A method of producing a composite solid polymer electrolyte membrane (SPEM) comprising a porous polymer substrate interpenetrated with an ion-conducting material, said method comprising the steps of solubilizing the ion-

conducting polymer and imbibing the porous polymer substrate with the ion-conducting polymer, and wherein

(i) the porous polymer substrate comprises a homopolymer or copolymer of a liquid crystalline polymer or a solvent soluble thermoset or thermoplastic aromatic polymer; and

(ii) the ion-conducting material comprises a homopolymer or copolymer of at least one of a sulfonated, phosphonated or carboxylated ion-conducting aromatic polymer or a perfluorinated ionomer.

86. (new) The method of claim 85, further comprising preparing a porous polymer substrate membrane, swelling of the porous substrate membrane with water, solvent exchanging the water swollen membrane and imbibing the solvent swollen polymer substrate membrane with the ion-conducting material.

87. (new) The method of claim 85, wherein the SPEM is substantially thermally stable to temperatures of at least about 100°C.

88. (new) The method of claim 85, wherein the SPEM is substantially thermally stable from at least about 100°C to at least about 175°C.

89. (new) The method of claim 85, wherein the liquid crystalline polymer substrate comprises a lyotropic liquid crystalline polymer.

90. (new) The method of claim 89, wherein the lyotropic liquid crystalline polymer substrate comprises at least one of a polybenzazole (PBZ) and polyaramid (PAR) polymer.

91. (new) The method of claim 90, wherein the polybenzazole polymer substrate comprises a homopolymer or copolymer of at least one of a polybenzoxazole

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(PBO), polybenzothiazole (PBT) and polybenzimidazole (PBI) polymer and the polyaramid polymer comprises a homopolymer or copolymer of a polypara-phenylene terephthalamide (PPTA) polymer.

92. (new) The method of claim 85, wherein the thermoset or thermoplastic aromatic polymer substrate comprises at least one of a polysulfone (PSU), polyimide (PI), polyphenylene oxide (PPO), polyphenylene sulfoxide (PPSO), polyphenylene sulfide (PPS), polyphenylene sulfide sulfone (PPS/SO₂), polyparaphenylene (PPP), polyphenylquinoxaline (PPQ), polyarylketone (PK) and polyetherketone (PEK) polymer.

93. (new) The method of claim 92, wherein the polysulfone polymer substrate comprises at least one of a polyethersulfone (PES), polyetherethersulfone (PEES), polyarylethersulfone (PAS), polyphenylsulfone (PPSU) and polyphenylenesulfone (PPSO₂) polymer; the polyimide (PI) polymer comprises a polyetherimide (PEI) polymer; the polyetherketone (PEK) polymer comprises at least one of a polyetherketone (PEK), polyetheretherketone (PEEK), polyetherketone-ketone (PEKK), polyetheretherketone-ketone (PEEKK) and polyetherketoneetherketone-ketone (PEKEKK) polymer; and the polyphenylene oxide (PPO) polymer comprises a 2,6-diphenyl PPO or 2,6 dimethyl PPO polymer.

94. (new) The method of claim 85, wherein the ion-conducting aromatic polymer comprises a wholly aromatic ion-conducting polymer.

95. (new) The method of claim 85, wherein the ion-conducting aromatic polymer comprises a sulfonated, phosphonated or carboxylated polyimide polymer.

96. (new) The method of claim 95, wherein the polyimide polymer is fluorinated.

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97. (new) The method of claim 94, wherein the wholly-aromatic ion-conducting polymer comprises a sulfonated derivative of at least one of a polysulfone (PSU), polyphenylene oxide (PPO), polyphenylene sulfoxide (PPSO), polyphenylene sulfide (PPS), polyphenylene sulfide sulfone (PPS/SO₂), polyparaphenylene (PPP), polyphenylquinoxaline (PPQ), polyaryleketone (PK), polyetherketone (PEK), polybenzazole (PBZ) and polyaramid (PAR) polymer.

98. (new) The method of claim 97, wherein:

(i) the polysulfone polymer comprises at least one of a polyethersulfone (PES), polyetherethersulfone (PEES), polyarylsulfone, polyarylethersulfone (PAS), polyphenylsulfone (PPSU) and polyphenylenesulfone (PPSO₂) polymer;

(ii) the polybenzazole (PBZ) polymer comprises a polybenzoxazole (PBO) polymer;

(iii) the polyetherketone (PEK) polymer comprises at least one of a polyetherketone (PEK), polyetheretherketone (PEEK), polyetherketone-ketone (PEKK), polyetheretherketone-ketone (PEEKK) and polyetherketoneetherketone-ketone (PEKEKK) polymer; and

(iv) the polyphenylene oxide (PPO) polymer comprises at least one of a 2,6-diphenyl PPO, 2,6-dimethyl PPO and 1,4-poly phenylene oxide polymer.

99. (new) The method of claim 85, wherein the perfluorinated ionomer comprises a homopolymer or copolymer of a perfluorovinyl ether sulfonic acid.

100. (new) The method of claim 99, wherein the perfluorovinyl ether sulfonic acid is carboxylic- (COOH), phosphonic- (PO(OH)₂) or sulfonic- (SO₃H) substituted.

101. (new) The method of claim 85, wherein the ion-conducting material comprises at least one of a polystyrene sulfonic acid (PSSA), poly(trifluorostyrene)

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sulfonic acid, polyvinyl phosphonic acid (PVPA), polyacrylic acid and polyvinyl sulfonic acid (PVSA) polymer.

102. (new) The method of claim 85, wherein the porous polymer substrate comprises a homopolymer or copolymer of at least one of a substituted or unsubstituted polybenzazole polymer, and wherein the ion-conducting material comprises a sulfonated derivative of a homopolymer or copolymer of at least one of a polysulfone (PSU), polyphenylene sulfoxide (PPSO) and polyphenylene sulfide sulfone (PPS/SO₂) polymer.

103. (new) The method of claim 102, wherein the polysulfone polymer comprises at least one of a polyethersulfone (PES) and polyphenylsulfone (PPSU) polymer.

104. (new) The method of claim 85, further comprising cross-linking the ion-conducting material to form sulfone crosslinkages.

105. (new) The method of claim 85, further comprising chlorinating or brominating the ion-conducting material.

106. (new) The method of claim 85, further comprising adding antioxidants to the ion-conducting material.

107. (new) The method of claim 85, further comprising purifying the ion-conducting material.

108. (new) The method of claim 107, wherein purifying the ion-conducting material comprises dissolving the ion-conducting material in a suitable solvent and precipitating the ion-conducting material into a suitable non-solvent.

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109. (new) The method of claim 107, wherein purifying the ion-conducting material comprises substantially removing overly sulfonated or degraded fractions of the ion-conducting material.

110. (new) A method of producing a composite solid polymer electrolyte membrane (SPEM) comprising a porous polymer substrate interpenetrated with an ion-conducting material, said method comprising the steps of preparing a polymer substrate and subsequently impregnating the polymer substrate with appropriate monomers which are then polymerized in-situ to form the composite SPEM, and wherein the porous polymer substrate comprises a homopolymer or copolymer of a liquid crystalline polymer or a solvent soluble thermoset or thermoplastic aromatic polymer.

111. (new) The method of claim 110, wherein the SPEM is substantially thermally stable to temperatures of at least about 100°C.

112. (new) The method of claim 110, wherein the SPEM is substantially thermally stable from at least about 100°C to at least about 175°C.

113. (new) The method of claim 110, wherein the liquid crystalline polymer substrate comprises a lyotropic liquid crystalline polymer.

114. (new) The method of claim 113, wherein the lyotropic liquid crystalline polymer substrate comprises at least one of a polybenzazole (PBZ) and polyaramid (PAR) polymer.

115. (new) The method of claim 114, wherein the polybenzazole polymer substrate comprises a homopolymer or copolymer of at least one of a polybenzoxazole

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(PBO), polybenzothiazole (PBT) and polybenzimidazole (PBI) polymer and the polyaramid polymer comprises a homopolymer or copolymer of a polypara-phenylene terephthalamide (PPTA) polymer.

116. (new) The method of claim 110, wherein the thermoset or thermoplastic aromatic polymer substrate comprises at least one of a polysulfone (PSU), polyimide (PI), polyphenylene oxide (PPO), polyphenylene sulfoxide (PPSO), polyphenylene sulfide (PPS), polyphenylene sulfide sulfone (PPS/SO₂), polyparaphenylene (PPP), polyphenylquinoxaline (PPQ), polyarylketone (PK) and polyetherketone (PEK) polymer.

117. (new) The method of claim 116, wherein the polysulfone polymer substrate comprises at least one of a polyethersulfone (PES), polyetherethersulfone (PEES), polyarylethersulfone (PAS), polyphenylsulfone (PPSU) and polyphenylenesulfone (PPSO₂) polymer; the polyimide (PI) polymer comprises a polyetherimide (PEI) polymer; the polyetherketone (PEK) polymer comprises at least one of a polyetherketone (PEK), polyetheretherketone (PEEK), polyetherketone-ketone (PEKK), polyetheretherketone-ketone (PEEKK) and polyetherketoneetherketone-ketone (PEKEKK) polymer; and the polyphenylene oxide (PPO) polymer comprises a 2,6-diphenyl PPO or 2,6 dimethyl PPO polymer.

REMARKS

The specification has been amended at pages 29 and 36 merely to reflect the deletions described above. Copies of original pages 29 and 36 which have been marked to show the respective revisions are enclosed herewith. Further, claims 1-50 have been cancelled and new claims 51-117 have been added. No new matter is presented by virtue of this Amendment. For example, support for new claims 51-117 appears throughout the specification, see e.g., pages 23 - 27, and in original claims 34-38.

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Applicants respectfully request entry of this Amendment prior to examination.

Early consideration and allowance of the application are earnestly solicited.

Respectfully submitted,

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